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Research Paper

Metamorphic consequences of secular changes in oceanic crust composition and implications for uniformitarianism in the geological record

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ABSTRACT

Cooling of the Earth's mantle since the Meso-Archean is predicted by thermal and petrological models to have induced a secular change in the composition of primary mantle-derived magmas – and thus bulk oceanic crust; in particular, suggesting a decrease in maficity over time. This hypothesis underpins several recent studies that have addressed key geological questions concerning evolving plate tectonic styles, the rates and timing of continental crust formation, comparative planetology, and the emergence of complex life on Earth. Major, minor, and trace element geochemical analyses of (meta)mafic rocks preserved in the geological record allows exploration of this theory, although no consensus currently exists about the magnitude of this change and what compositions – if anything – constitute representative examples of Paleo-, Meso-, or Neo-Archean primitive oceanic crust. In this work, we review the current state of understanding of this issue, and use phase equilibria to examine the different mineral assemblages and rock types that would form during metamorphism of basalt of varying maficity in subduction zone environments. The presence (or absence) of such metamorphic products in the geological record is often used as evidence for (or against) the operation of modern-day subduction-driven plate tectonics on Earth at particular time periods; however, the control that secular changes in composition have on the stability of mineral assemblages diagnostic of subduction-zone metamorphism weakens such uniformitarianistic approaches. Geodynamic interpretations of the Archean metamorphic rock record must therefore employ a different set of petrological criteria for determining tectonothermal histories than those applied to Proterozoic or Phanerozoic equivalents.

1. Introduction

"By explaining past changes by analogy with present phenomena, a limit is set to conjecture, for there is only one way in which two things are equal, but there is an infinity of ways in which they could be supposed different." – Hooykaas (1963).

Basalt is the most widespread rock on the Earth's surface today (Nielsen and Fisk, 2010), and investigations of Phanerozoic, Proterozoic, and Archean terranes worldwide suggest that it has been similarly abundant throughout geological time (Condie, 1981; Furnes et al., 2014). As oceanic crust is produced via decompression melting of mantle peridotite at divergent plate margins (McKenzie and Bickle, 1988), its structure and composition can provide critical first-order constraints on the geochemical and petrophysical properties of the Earth's interior (Vervoort and Blichert-Toft, 1999). Even basalts produced in intraplate geodynamic scenarios, such as above the heads of mantle plumes, can provide critical information about the thermal and petrological properties of the Earth's shallow interior (Campbell and Griffiths, 1990). Identifying primary magmas or otherwise relatively unaltered components of oceanic crust in the geological record is thus of prime importance, as such discoveries would help to resolve many major issues in solid-Earth geoscience, such as constraining the thermal evolutions of the mantle over time and deducing whether or not plate tectonics operated during the Archean (e.g. Davies, 1992; Hamilton, 1998, 2003; Stern, 2005; Van Hunen and Moyen,

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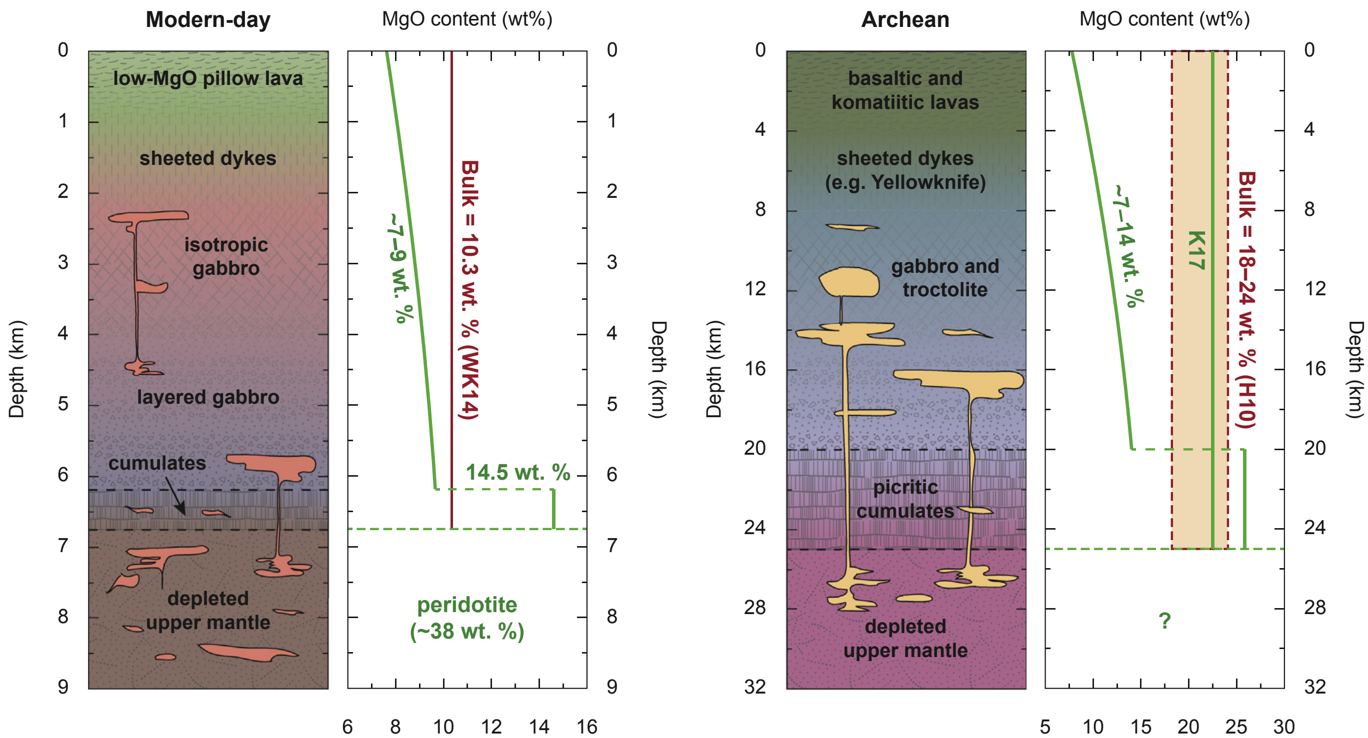

Figure 1. Schematic cross sections through modern-day and (interpreted) Archean oceanic crust and upper mantle. Modern-day structure, petrology, and MgO contents are from White and Klein (2014), and Archean structure and petrology are after Foley et al. (2003) and Nair and Chacko (2008). Bulk MgO content for Archean crust is after Herzberg et al. (2010) (H10), and two compositional profiles discussed by Klein et al. (2017) (K17) and in this study, are shown by thick green lines.

2012; Turner et al., 2014; Ernst, 2017; Ganne and Feng, 2017; Wade et al., 2017).

Continued improvements to quantitative chemical analysis techniques in recent years—alongside advances in large-scale data archiving and distribution—have heralded the rise of Big Data within the geological community (Kattge et al., 2014). Centralized, public-access repositories containing an extensive range of petrological analyses of rocks and minerals (e.g. EarthChem[1]) allow straightforward processing of vast amounts of information. Indeed, several recent studies have analyzed large databases of Proterozoic and Archean metabasalt bulk-rock compositions to examine the extent of secular variations in major, minor, and trace element geochemistry, and elucidate the geodynamic settings of formation of the parental mafic magmas (Keller and Schoene, 2012; Furnes et al., 2014; Condie et al., 2016; Ganne and Feng, 2017). These studies, however, have produced differing opinions on the extent to which the petrology and structure of primary mafic oceanic crust and/or mid-ocean ridge basalt (MORB) has evolved over geological time, which are predicted by models of the mantle's thermal history to be significant (Abbott et al., 1994; Korenaga, 2008; Herzberg et al., 2010; Ziaja et al., 2014).

Nearly all basalts in the Precambrian rock record have undergone some degree of metamorphism (Gill, 1979), sometimes in subduction zones at high-pressure/low-temperature (HT–LP) conditions, or alternatively in collisional orogens along notably hotter geothermal gradients (e.g. Brown, 2007). Secular changes in primary mantle-derived magmas, bulk oceanic crust, and/or MORB compositions will thus have distinct petrological consequences on the metamorphic rock type produced in each tectonic setting (e.g. Palin and White, 2016). Thus, there is a need to define the mineral parageneses that are expected to form in different mafic rocks at these variable *P*–*T* conditions, which will allow more informed evaluation of the rock record and can provide greater insight into how metamorphic processes and products may have evolved throughout Earth history (e.g. Grambling, 1981; Sandiford, 1989; Komiya et al., 2002; Martin and Moyen, 2002; Brown, 2007; Bradley, 2011; Zhai and Santosh, 2013; Dyck et al., 2015; Weller and St-Onge, 2017; Nicoli and Dyck, 2018).

In this paper, we first summarize the theory behind secular change in bulk oceanic crust and MORB composition, and discuss the extent to which metabasic rocks preserved in the geological record validate these predictions. We then present the results of phase diagram analysis and interpretation demonstrating the petrological consequences of such compositional changes at *P*–*T* conditions characteristic of modern-day, and early and late Archean subduction zones. Finally, we discuss the implications of this theoretical secular change on uniformitarianism-based approaches to interpreting the geological record, and the onset of modern-day subduction-driven plate tectonics.

1.1. Predicted secular change in bulk oceanic crust and uppermost MOR basalt compositions

It has long been known that the Earth has undergone cooling since its formation (Thomson, 1862; See, 1907; Patterson, 1956); however, the absolute magnitude and rate of this temperature change over time is poorly constrained. Conceptual models for this thermal evolution fundamentally rely upon the balance at any point in time between internal heat production from the decay of radiogenic elements, and the loss of heat by mantle convection and surface volcanism (Breuer and Spohn, 1993). The present-day ratio of heat production to heat loss—the convective Urey ratio—is estimated to be 0.23 ± 0.15 (Korenaga, 2008), and can be

[1] http://www.earthchem.org/.

extrapolated back in time based on constraints provided by geophysical and geochemical models of the Earth (e.g. Turcotte, 1980). Such an exercise generally predicts a concave-upwards thermal-evolution curve for the Earth's mantle potential temperature (T_P) that peaks in the Meso-Archean (~2.8–3.2 Ga), inferring an ambient upper mantle that was around 300 °C hotter than today's value of ~1350 °C (Korenaga, 2006; Herzberg et al., 2010). These data imply an average mantle cooling rate of ~100 °C/Ga since this time.

Primary oceanic crust produced at spreading centers on Earth today is ~6–7 km thick (Fig. 1), has an average bulk MgO content of 10–13 wt.%, and forms from small mantle melt fractions of 8–10% (Herzberg et al., 2010). The uppermost portion of the mafic crust (i.e. MORB) is typically comprised of pillow basalts with an MgO content of ~7 wt.% (White and Klein, 2014). A necessary petrological result of a hotter Archean mantle T_P up to 1650 °C is deeper and more voluminous melting of peridotite (25–45% melt fraction), which likely produced a thicker (~25–40 km) crust (McKenzie and Bickle, 1988) with a greater average bulk MgO content of ~18–24 wt.% (Fig. 1; Abbott et al., 1994; Van Thienen et al., 2004; Herzberg et al., 2010). Geochemical models of Archean primary magmas based on theoretical mantle protolith compositions, T_P values, and higher melt fractions predict that the uppermost, MORB-like portions of this primitive crust would have MgO contents in the range ~11–15 wt.% (Ziaja et al., 2014). Secular cooling of mantle T_P over time would thus suggest that the maficity of mantle-derived basalts has decreased throughout geological time, although a range of non-equilibrium processes (e.g. fractional crystallization) may act to alter the mineralogy and composition of the final crystallized product.

1.2. Observations from the geological record

The Archean rock record is represented by 35 fragments of continental lithosphere – or cratons (Bleeker, 2003) – that cover just ~4.7% of the Earth's surface (Artemieva, 2006). Such regions are typically characterized by sodic granitoids of tonalite–trondhjemite–granodiorite (TTG) composition (Jahn et al., 1981; Moyen and Martin, 2012; White et al., 2017), and mafic-to-ultramafic volcanic rocks and sediments, most of which are typically metamorphosed to greenschist-facies *P–T* conditions (Condie, 1981). These "greenstone" terranes have been suggested by some workers as representing – or else containing – variably altered remnants of primary Archean oceanic crust, supported in some cases by modern-day, ophiolite-like macrostructures including pillow lavas and sheeted dykes (e.g., Helmstaedt et al., 1986; Kusky and Kidd, 1992; Furnes et al., 2007; Grosch and Slama, 2017). However, many greenstone terranes may be autochthonous (Bickle et al., 1994; Thurston, 2002), possibly representing continental rift or flood basalts (Bleeker, 2002), or formed in a non-MOR marine environment (i.e. arc- or plume-related), as inferred by incompatible trace-element ratios characteristic of different modern-day mantle source regions (Pearce, 2008). As such, not all (meta)basalts can be used to assess the existence or not of secular compositional change in primary crust composition from the Archean to the modern day.

Early studies of geochemical variations between modern-day and Archean basalts focused mostly on differences in trace element ratios, and either made comparisons when bulk-rock data were normalized to a common Mg# [= 100 × Mg/(Mg + Fe^{2+})] (e.g. Gill, 1979) or applied arbitrary compositional limits to eliminate highly evolved or highly fractionated lithologies from consideration, such as pyroxene- or olivine-rich cumulates (e.g. Condie, 1985). More recent works have taken advantage of Big Data to allow more extensive spatial and temporal coverage of metabasalts worldwide, and have utilized more in-depth statistical tests to ensure significance of results (e.g. Ganne and Feng, 2017). Four major studies have been performed in recent years, with all having identified secular changes in the MgO content (and other chemical components) of basalt to different degrees (Table 1).

Table 1
Reported secular trends in oceanic crust compositions since the early Archean. Only elements reported as showing secular variation are noted here: if absent from this table, they were either reported to have remained approximately constant over time, or were not discussed. N.B. Mg# [= 100 × Mg/(Mg + Fe^{2+}) in cation units], [1]basalts with depleted mantle trace element geochemistry.

Lithology	Increase	Decrease	Study
Tholeiitic basalt	Al, Ti, Zr, P	Cr, Ni, Co	Gill (1979)
Tholeiitic basalt	Al	Cr, Ni, Co	Condie (1985)
Basalt	K, Na	Mg, Cr, Ni	Keller and Schoene (2012)
Basalt/greenstone	Al, Ti, Zr	Mg, Ni	Furnes et al. (2014)
Depleted mantle basalt[1]	Na, Ti, Mg#	Fe, Mn	Condie et al. (2016)
Archean basalt	Al	Mg	Ganne and Feng (2017)

Furnes et al. (2014) and Condie et al. (2016) analyzed geochemical databases of metabasalt compositions obtained from over 100 Precambrian greenstone belts worldwide, and used incompatible trace element ratios to determine probable geodynamic environments of formation for each locality. From these data, Furnes et al. (2014) reported a relatively large, statistically significant decrease in bulk-rock MgO content from ~15 wt.% at 3.5 Ga to ~7 wt.% today, although Condie et al. (2016) reported a smaller decrease, with a mean MgO content of ~9 wt.% for rocks older than 3 Ga and ~7 wt.% for younger samples. However, neither study accounted for spatiotemporal data clustering within their respective databases, indicating that calculated average compositions and trend magnitudes may have been influenced by sampling bias to unknown (but potentially non-trivial) degrees.

In order to account for sample bias, Keller and Schoene (2012) and Ganne and Feng (2017) produced weighted databases of samples from the mafic magma record, which were then analyzed via a Monte Carlo resampling procedure designed to minimize selection bias in space and time. While Keller and Schoene (2012) reported a clear secular decrease in MgO content for the global geochemical average mafic rock, they did not discriminate between possible environments of formation for each sample, meaning that this reported trend lacks key geodynamic context needed to support a specific change in primitive basalt/oceanic crust composition over time. By contrast, Ganne and Feng (2017) filtered out plutonic and hypabyssal units to distinguish rocks derived directly from mantle melting from those affected by subsequent evolution and fractionation processes in the lithosphere or continental crust. Further, no preselection of data via different trace element ratios was applied, to minimize the chance of introducing unintentional selection bias based on incorrectly translating modern-day definitions to early-Earth geodynamic environments (cf. Li et al., 2015). From this reduced dataset, provided in a supplementary database, Ganne and Feng (2017) reported successful primary magma solutions for Archean lithologies that had an average MgO content of ~20 wt.%, and equivalent interpreted basalt compositions with a mean MgO content of ~13 wt.%. Although it is not possible to show that these Archean lavas formed in an ocean-ridge setting, this high-MgO content exceeds that reported for modern-day MORB (White and Klein, 2014), and agrees with reports of strongly mafic pillow lavas (>12 wt.%) with MOR-like geochemical affinity that occur in some Paleo-Archean greenstone belts (e.g. Isua; Komiya et al., 2002, 2004).

2. Petrological implications of secular compositional changes

The secular changes in primary basalt composition predicted by thermal modeling and observed in the geological record indicate

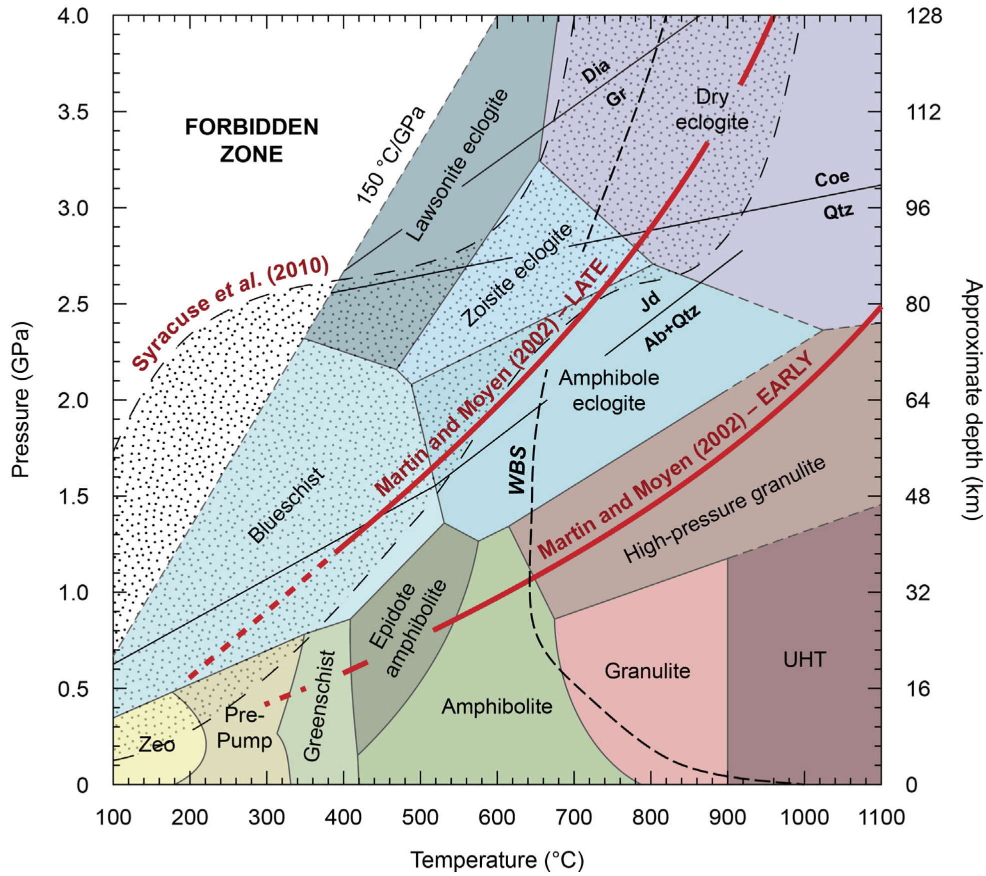

Figure 2. Classical metamorphic facies diagram (modified after Maruyama et al., 1996) overlain by pressure–temperature (*P*–*T*) profiles (red lines) reported from the literature to characterize early and late Archean subduction channels (cf. Martin and Moyen, 2002). The late Archean subduction-zone geotherm is approximately 500 °C/GPa and the early Archean subduction-zone geotherm is approximately 350 °C/GPa. Dotted band represents the range of *P*–*T* conditions calculated for descending slab surfaces in all modern-day subduction zones (Syracuse et al., 2010). Geotherm marked 150 °C/GPa denotes the limit of the Forbidden Zone (Liou et al., 2000). Abbreviations for facies and minerals are as follows: Zeo = zeolite, Pre-Pump = prehnite–pumpellyite, UHT = ultrahigh temperature, Dia = diamond, Gr = graphite, Qtz = quartz, Coe = coesite, Jd = jadeite, Ab = albite. Water-saturated basalt solidus (WBS) is after Vielzeuf and Schmidt (2001) and Palin et al. (2016b).

the importance of characterizing the metamorphic products of variably magnesian rocks that occur in all greenstone terranes worldwide (e.g. Kato and Nakamura, 2003). Although Earth's first continents likely formed in intraplate tectonic settings in a so-called plume-lid or stagnant-lid geodynamic regime (e.g. Smithies, 2000; Bédard, 2006; Van Kranendonk, 2010; Debaille et al., 2013; Palin et al., 2016c), there are multiple independent lines of evidence suggesting that subduction had begun to operate on a global scale by the Meso-Archean, as detailed below. As this point in time coincides with the reported peak in mantle T_P (Herzberg et al., 2010), and so the most MgO-rich mafic crust, it is pertinent to examine the petrological products of subducted and metamorphosed oceanic crust of variable maficity. As such, we have focused our petrological analysis on the parageneses that form at *P*–*T* conditions representative of subduction zone environments on the modern-day Earth and the early and late Archean. We have considered a diverse set of protolith bulk compositions—both natural and synthetic—that encompasses the expected range of maficity reported for bulk oceanic crust and MORB throughout geological time.

2.1. Early-Earth plate velocities and subduction geotherms

Although there is long-standing debate within the geological community regarding the timing of initiation of plate tectonics (e.g. Hamilton, 2003; Stern, 2005; Condie and Kröner, 2008; Hopkins et al., 2008; Turner et al., 2014; Maruyama et al., 2017), it is generally thought that subduction had begun to operate on Earth on a global scale by ∼3.0 Ga (cf. Shirey and Richardson, 2011; Korenaga, 2013; Nutman et al., 2015). We adopt this interpretation, as it has the advantage of providing a consistent geodynamic framework with which to investigate the petrological consequences of secular compositional changes, although increased buoyancy of Archean oceanic lithosphere may have promoted shallower subduction than commonly occurs today (Van Hunen and Moyen, 2012). Petrological arguments for and against subduction having operated at this time are addressed in section 4 of this paper within the context of our petrological calculations.

The geometries and thermal structures that may have characterized Archean subduction zones are unknown, despite both being critical factors for determining the metamorphic and magmatic rock types that form there (e.g. Peacock; 1996; Van Keken et al., 2002). Thermo-mechanical numerical modeling of heat transfer in subduction zones has shown that the *P*–*T* evolutions of various parts of the subducting lithosphere are primarily controlled by plate velocity, slab age, and dip angle (Kirby et al., 1991), although additional criteria such as mantle T_P, heat capacity, and the nature of phase transitions also play a role (Peacock and Wang, 1999; King, 2001; Gerya et al., 2002, 2004; Billen, 2008; Palin et al., 2017). While studies have shown that calculated slab-surface *P*–*T* conditions are most sensitive to plate velocity, slab age, and dip angle, which can vary widely in natural geological systems (Syracuse

Table 2
Bulk compositions of lithological components discussed in this work (wt.% oxide). [1]sample CHK-MGO14; [2]sample 485418; [3]sample 02MB256. N.B. MAOC = petrological model of Archean oceanic crust with 11, 13, and 15 wt.% MgO. In all studies, measured Fe is reported here as FeO.

Lithology	Reported age	SiO_2	TiO_2	Al_2O_3	FeO	MgO	CaO	Na_2O	K_2O	Study
Bulk oceanic crust	Modern day (<200 Ma)	50.10	1.10	15.70	8.30	10.30	11.80	2.21	0.11	White and Klein (2014)
	"Archean" (MAOC11)	48.50	0.72	16.65	8.13	11.00	12.78	1.71	–	Ziaja et al. (2014)
	"Archean" (MAOC13)	48.14	0.69	15.81	8.12	13.00	12.14	1.63	–	Ziaja et al. (2014)
	"Archean" (MAOC15)	47.78	0.65	14.97	8.11	15.00	11.50	1.54	–	Ziaja et al. (2014)
Modern MORB	Modern day (<200 Ma)	50.06	1.52	15.00	10.36	7.71	11.46	2.52	0.19	White and Klein (2014)
Olivine-bearing basalt[1]	Paleoproterozoic (ca. 1870 Ma)	49.96	0.79	11.00	10.97	14.07	11.57	1.30	0.09	Hynes and Francis (1982)
High-MgO pillow basalt[2]	Mesoarchean (ca. 3075 Ma)	48.41	0.43	10.87	11.26	14.90	11.26	1.23	0.15	Polat et al. (2008)
Komatiitic basalt[3]	Paleoarchean (ca. 3450 Ma)	48.75	0.48	10.32	10.90	16.19	10.49	1.42	0.03	Kato and Nakamura (2003)

et al., 2010; Van Hunen and Moyen, 2012; Sizova et al., 2015), various assumptions can be made about these criteria in the Archean from many independent lines of evidence.

Early studies of Archean geodynamics favored the idea that plate tectonics operated at a faster rate than on Earth today (Hargraves, 1986), given that the dissipation of internal heat from a significantly hotter mantle would be most efficient in a scenario involving rapid sea-floor spreading and subduction of relatively young (20–30 Myr) crust (e.g. Bickle, 1978; Abbott and Hoffman, 1984). Archean spreading rates up to ~40 cm/yr were proposed by some workers (e.g. Dewey and Windley, 1981; Nisbet and Fowler, 1983), which are significantly higher than present-day rates of 1–10 cm/yr (Solomon et al., 1975). While some workers continue to support this 'fast-plate' hypothesis (Van Kranendonk and Kirkland, 2013), Archean plate velocities are now thought to have been more sluggish than today (Korenaga, 2013), as indicated by paleomagnetic and paleogeographic data (Condie, 2015), the geochemical budget of isotope exchange between the hydrosphere and Earth's interior (Padhi et al., 2012), calculated constraints on the Urey ratio over time (Korenaga, 2008), and passive margins having had longer lifespans in the Precambrian than in the Phanerozoic (Bradley, 2008).

Quantitative estimates of Archean plate velocities based on a model of time-dependent mantle heat flux were presented by Korenaga (2006). When scaled against an average present-day value of 4 cm/yr, the Proterozoic–Archean boundary (2.5 Ga) was characterized by a velocity of ~3 cm/yr, and the Archean–Hadean boundary (4 Ga) by ~2 cm/yr. In addition, subducted crust was likely to have been older and colder (Korenaga, 2013) than the ~60-Myr present-day average (Bickle, 1978). The effect that slower and older descending crust may have on subduction zone thermal structure can be interpreted on a first-order basis via comparison with present-day analogues that show similar physical parameters, such as the Lesser Antilles, where subducting slab surfaces have P–T profiles with geotherms of ~300–400 °C/GPa (Syracuse et al., 2010).

Alongside these theoretical arguments, first-order evidence of relatively cool subduction during the Meso–Archean (ca. 3.2 Ga, Moyen et al., 2006; and ca. 2.87 Ga, Mints et al., 2010) and Paleo-Proterozoic (ca. 2.2–1.8 Ga; Ganne et al., 2011; Weller and St-Onge, 2017) is recorded within high-pressure metasediments and metabasites preserved in the geological record. In these works, conventional thermobarometry and phase equilibrium modeling results indicated peak subduction-related metamorphism along geotherms of ~350–400 °C/GPa, which are indeed similar to most modern-day slab-top P–T profiles (Fig. 2). As some geodynamic models suggest that increased lithospheric buoyancy in the early Archean caused subduction to have operated at shallower angles than in the late- and post-Archean (Van Hunen and Moyen, 2012; Sizova et al., 2014), we have considered changes in phase assemblages predicted to be stable in a range of basaltic compositions along both the warm early-Archean (~600 °C/GPa) and cold late-Archean (~300 °C/GPa) subduction zone geotherms of Martin and Moyen (2002) (Fig. 2).

2.2. Calculated metamorphic parageneses

Mafic igneous rocks and their metamorphosed equivalents show most compositional variability within the MnO–Na_2O–CaO–K_2O–FeO–MgO–Al_2O_3–SiO_2–H_2O–TiO_2–Fe_2O_3–CO_2 (MnNCKFMASHTOc) chemical system; however, first-order interpretations of key phase changes need only consider Na_2O–CaO–FeO–MgO–Al_2O_3–SiO_2–H_2O (e.g. Spear, 1995). For projection and visualization purposes, further reduction into the ACF ($AlO_{3/2}$–CaO–FeO + MgO) ternary compositional system was performed here using Perple_X (Connolly, 2009) and the thermodynamic dataset of Holland and Powell (2011). A high activity of H_2O (i.e. free fluid) was assumed for metamorphism at subsolidus conditions (Schmidt and Poli, 1998) alongside the occurrence of minor quartz or coesite (SiO_2), depending on pressure. While modern-day petrological techniques can produce bulk-rock specific equilibrium phase assemblage diagrams (pseudosections) for metamafic rocks (Powell et al., 1998; Green et al., 2016), these calculated equilibria are often distinctly sensitive to small variations in bulk composition oxide ratios (e.g. Stüwe, 1997; Guevara and Caddick, 2016; Palin et al., 2016a) and complexities in the stabilities of minor phases can mask broader-scale – and more important – petrological features. Indeed, as no one particular bulk composition can be shown to be completely representative of primary mafic oceanic crust/MOR basalt at any point in the geological past (section 1.2), we approach the problem by investigating the effects that general compositional trends in the major varying components MgO and Al_2O_3 (Table 1) have on first-order changes in metamorphic mineral assemblages. We note that reported FeO contents of mafic crust have remained generally constant throughout geological time (e.g. Condie et al., 2016; Table 1), meaning that changes in MgO content itself lead to variation in overall bulk-rock Mg#.

All bulk-rock compositions considered herein (Table 2) are shown in ACF compositional space in Fig. 3a, alongside common minerals that form in metabasic rocks in subduction zone environments (e.g. Rebay et al., 2010; Maldonado et al., 2016). Observed modern-day (White and Klein, 2014) and calculated Archean (MAOC: Ziaja et al., 2014) bulk oceanic crust compositions are shown in Fig. 3b for reference, and a series of natural basalts of Neo-Proterozoic to Paleo-Archean age (Table 2) of varying maficity are shown in Fig. 3c, which we use to examine the petrological effects of reported secular change in MORB composition. This uppermost basaltic portion of the oceanic crust – MORB – is specifically examined here as it is likely to be hydrated during crust formation, seafloor metasomatism (Peck et al., 2001), or lithospheric deformation at plate boundaries (Korenaga, 2017), and so undergoes fluid-mediated equilibrium metamorphism during subduction (Guiraud et al., 2001). These natural samples were selected from

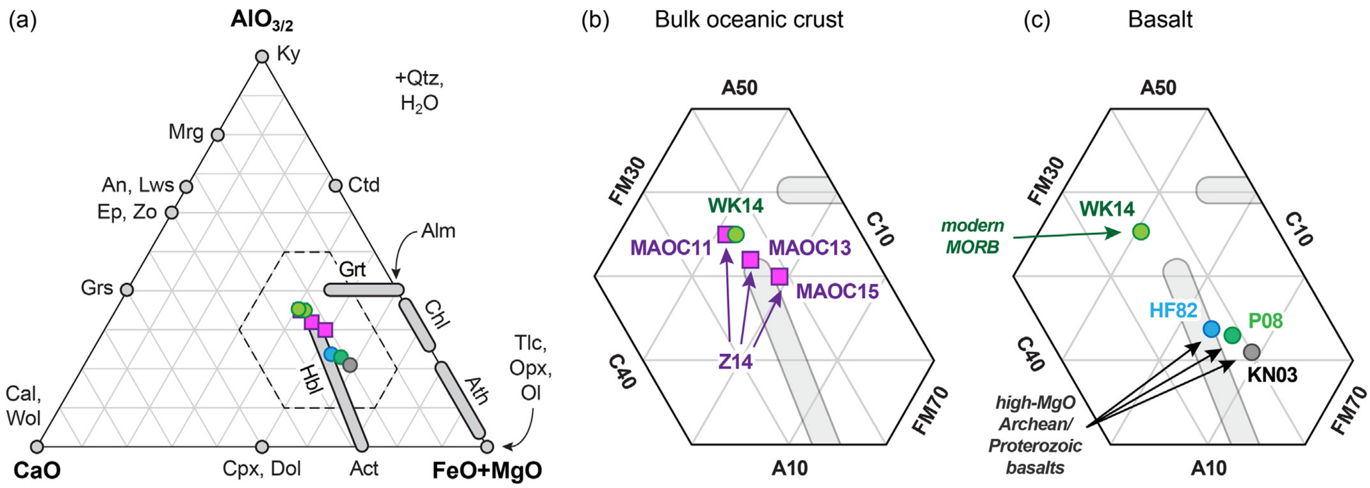

Figure 3. ACF ($AlO_{3/2}$–CaO–FeO + MgO) chemographic projections showing the compositions of mafic oceanic crustal lithologies of various ages. End-member mineral abbreviations are after Kretz (1983). (a) ACF diagram showing minerals commonly observed in metamorphosed mafic rocks alongside each bulk composition discussed in the text and Table 2. (b) Magnification of part (a), showing the position in ACF space of estimated compositions of bulk oceanic crust today (WK14: White and Klein, 2014) and in the Archean (Z14: Ziaja et al., 2014). N.B. MAOC = petrological model of Archean oceanic crust with 11, 13, and 15 wt.% MgO. (c) Magnification of part (a) showing the position in ACF space of basalt compositions of early Archean to modern-day age. HF82 = Hynes and Francis (1982), KN03 = Kato and Nakamura (2003), P08 = Polat et al. (2008), WK14 = White and Klein (2014). See Table 2 for full compositions.

greenstone terranes identified by Furnes et al. (2014) as having MOR affinity. Average modern-day MORB (White and Klein, 2014) is also shown on Fig. 3c for comparative purposes. All mineral abbreviations are after Kretz (1983), including Ab = albite, Jd = jadeite, Qtz = quartz, and H_2O = water as projected phases. While quartz may not always be present in metamorphosed ultramafic rocks, examination of phase relations in ACF compositional space is valid if silica (SiO_2) is a component in associated projected phases, such as albite or jadeite, and if all phases on the projection plane contain it (cf. Green et al., 1968).

A series of ACF compatibility triangles showing equilibrium phase assemblage changes at *P*–*T* conditions along an early Archean warm geotherm (~600 °C/GPa; Fig. 2) are shown in Fig. 4a–d. The bulk compositions reported in Table 2 and shown in Fig. 3c may be separated into low-MgO modern-day MORB (Mo-MORB) and high-MgO Archean–Proterozoic basalts (High-Mg APB). At relatively low-grade epidote-amphibolite facies conditions (500 °C, 0.8 GPa), low-MgO protoliths would be characterized under metamorphic conditions by chlorite-, zoisite-, and actinolite-rich assemblages, whereas basalts with higher MgO contents would be zoisite-free. During prograde metamorphism, zoisite is replaced by plagioclase feldspar and/or lawsonite during the transition into lower amphibolite-facies *P*–*T* conditions, with garnet potentially forming in Al_2O_3-rich and MgO-poor lithologies (Fig. 4b). With increasing metamorphism and subduction to greater depths, transition to upper amphibolite/lower granulite-facies conditions (700 °C, 1.2 GPa) produces plagioclase/lawsonite–garnet–hornblende assemblages in low-MgO protoliths, and garnet–hornblende–cummingtonite/orthopyroxene assemblages in high-MgO protoliths (Fig. 4c). The garnet–hornblende tie line that forms in ACF compositional space distinctly separates these two protolith groups, although pressures representative of this geotherm are insufficient to destabilize amphibole and thus produce eclogite (*sensu stricto*) or hornblende-free two-pyroxene granulite in these bulk compositions. However, at granulite-facies conditions of >800 °C and >1.5 GPa, the garnet–hornblende tie line is replaced by the garnet–clinopyroxene tie line (Fig. 4d), suggesting that low-MgO protoliths would produce partially melted high-pressure granulite during Archean subduction, but high-MgO protoliths would instead produce garnet pyroxenite. Such pyroxenitic lithologies are rare in Archean cratons (Bédard, 2017), and so – if formed on the early Earth – likely terminally descended into the aesthenospheric mantle as non-exhumable crust (Bédard, 2006).

A similar petrographic treatment is applied to theoretical subduction along a relatively cool geotherm (~300 °C/GPa; Fig. 2), thought to be representative of late-/post-Archean slab-top *P*–*T* paths (Fig. 4e–h) (Martin and Moyen, 2002). At 500 °C and 1.6 GPa along this geotherm, representative of the earliest stages of subduction, all considered bulk-rock compositions fall within a quadrilateral defined by zoisite, garnet, clinopyroxene, and sodic amphibole (Fig. 4e). At lower blueschist-facies conditions, this four-phase assemblage is divided by a zoisite–glaucophane tie line, below which all considered bulk-rock compositions lie (Fig. 4e). At higher temperature conditions representative of the upper blueschist facies, a tie-line switch occurs stabilizing garnet–clinopyroxene assemblages in place of zoisite–glaucophane assemblages. At this point, low-MgO bulk compositions become separated in ACF compositional space from high-MgO bulk compositions, with the former containing zoisite and the latter containing sodic amphibole (Fig. 4e). At higher-grade conditions along this geotherm (600 °C, 2 GPa), representative of the amphibole-eclogite facies of Maruyama et al. (1996), all bulk compositions considered are predicted to contain eclogitic parageneses (garnet and clinopyroxene), with or without zoisite, glaucophane, and talc (Fig. 4f). Similar equilibrium assemblages occur at higher grade (750 °C, 2.6 GPa; Fig. 4g), with the sole difference pertinent to the rock types considered being the stabilization of kyanite in high-Al_2O_3/low-MgO bulk compositions. No significant changes in ACF phase equilibria occur during deep subduction to ultrahigh-pressure "dry eclogite"-facies conditions (>800 °C, >2.9 GPa; Fig. 4h), aside from the transformation of quartz to coesite and likely loss of free H_2O due to prograde devolatilization. All but the most magnesian protolith (komatiitic basalt) are predicted to transform to eclogite (*sensu stricto*), while these near-ultramafic precursor basalts would instead transform to garnet pyroxenite (Fig. 4h; Johnson et al., 2014), as also predicted by experimental petrology (Foley et al., 2003; Ziaja et al., 2014). Metamorphosed olivine-normative basalt is unlikely to form quartz/coesite at eclogite-facies conditions, with previous calculations predicting its presence during subduction up to *P* ~22 kbar (Palin and White, 2016); however, metamorphosed

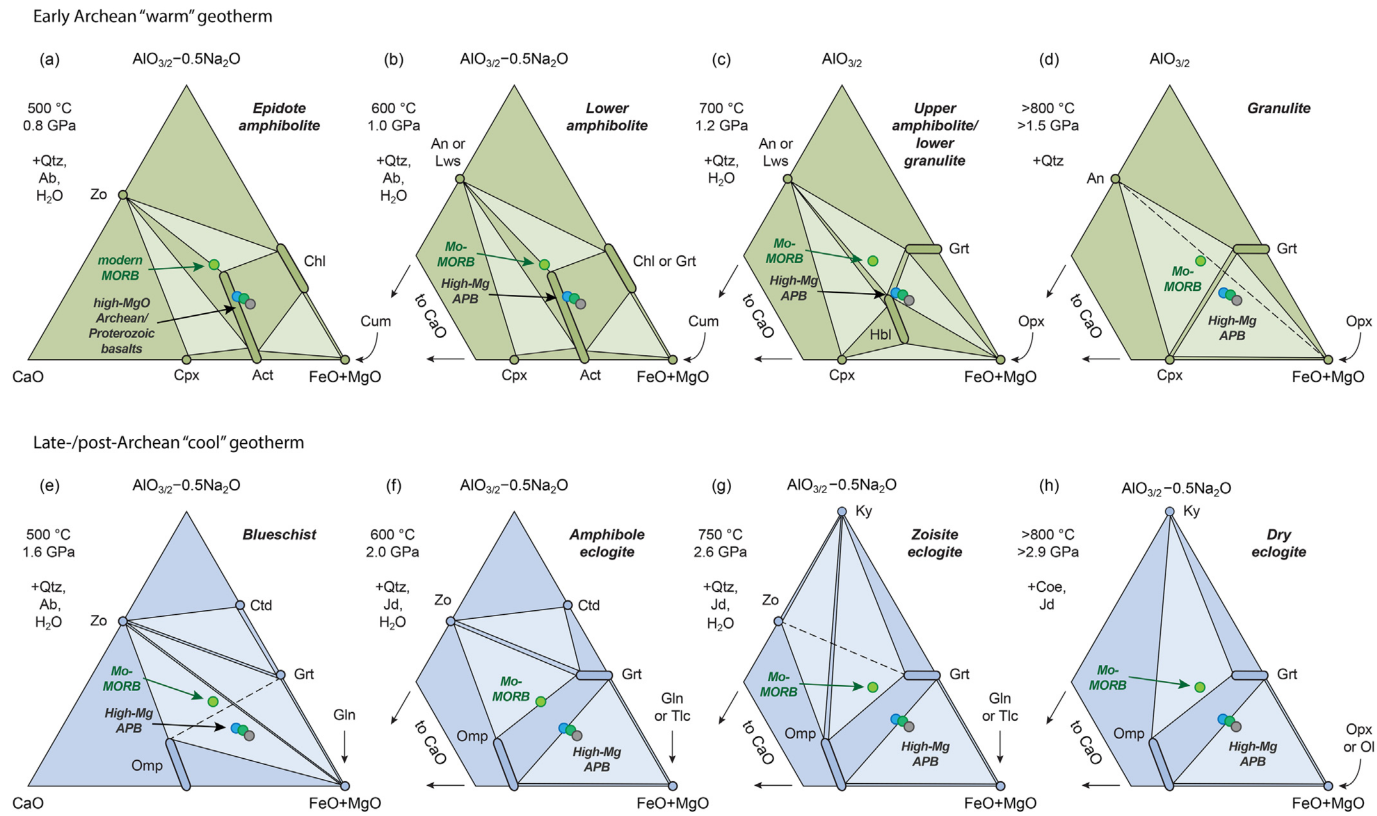

Figure 4. ACF chemographic projections showing phase equilibria at pressure–temperature (P–T) conditions along the (a–d) early-Archean "warm" geotherm (600 °C/GPa) and (e–h) late-/post-Archean "cool" geotherm (300 °C/GPa) of Martin and Moyen (2002). End-member mineral abbreviations, including those in projection, are after Kretz (1983). Bulk compositions of modern-day and Archean/Proterozoic basalts are shown for reference. The compositions used for ACF plots are those shown in Fig. 3.

quartz-normative tholeiitic basalts commonly contain either polymorph at dry eclogite conditions (Fig. 2; St-Onge et al., 2013).

3. Discussion

Determining whether components of oceanic crust preserved in the geological record show time-dependent evolutions in bulk composition is critical to validating predictions made by thermal and petrological models (e.g. Korenaga, 2006). Such *prima facie* evidence of secular trends would also have immense value in bounding changes in key—yet poorly constrained—whole-Earth thermal parameters over time, such as the convective Urey ratio and mantle T_P (Herzberg et al., 2010; Ganne and Feng, 2017). However, despite extensive field investigations, no coherent oceanic crustal sections/ophiolites have been documented from Archean terranes (Bickle et al., 1994), and some reported occurrences have since been disputed by other members of the community (e.g. Furnes et al., 2007; Nutman and Friend, 2007), and so no primary constraints exist on the extent to which early-Earth oceanic crust was structurally or compositionally stratified (cf. Fig. 1).

3.1. Secular change in MOR basalt composition: fact or fiction?

While theoretical arguments for a secular change in the overall maficity of bulk oceanic crust composition are robust (section 1.1), it is still uncertain whether upper-oceanic crust basalts in the Archean were commensurately more MgO-rich than modern-day MORB. Several workers have examined this issue and present conflicting conclusions about the magnitude of change over time. While Keller and Schoene (2012), Furnes et al. (2014), and Ganne and Feng (2017) documented significant decreases in global average (meta)basalt maficity since the Meso-Archean, which approached ~15 wt.% MgO at 3.5 Ga, Condie et al. (2016) reported a lesser decrease of just ~2 wt.% over the same period. If MORB has retained a more-or-less constant MgO content throughout geological time, mass-balance constraints require that the lowermost portions of Archean oceanic crust must have been significantly more magnesian than modern-day examples (Fig. 1), as the overall bulk-crustal MgO content was much higher. However, no such rocks in the geological record are thought to be definitive examples of these lower-crustal cumulate lithologies (Bickle et al., 1994). By contrast, some authors suggest that Archean oceanic crust may have consisted predominantly of komatiite-like high-MgO basalts, with no significant vertical stratification in composition or structure (Fig. 1) (Arndt, 1983; Klein et al., 2017), with the low-MgO basalts found in many greenstone belts (Condie and Aster, 2009) potentially having been derived via burial metamorphism and partial melting of these original high-MgO picritic/komatiitic protoliths. If true, this supports the hypothesis that petrological indicators of subduction – as defined by assemblages that stabilize in modern-day, low-MgO MORB – would not have formed on the early Earth, even if identical metamorphic P–T conditions were reached (Palin and White, 2016).

If each of the four main studies discussed herein have used largely similar geochemical datasets, why is such significant variation in compositional change over time be predicted? One prominent issue may relate to how 'non-arc' or 'MOR-derived basalt' is defined in each case. If all mafic rocks in greenstone belts are used for statistical analysis of secular compositional change, a major decrease in global average MgO content with time is observed, with a significant step occurring at ~2.5 Ga (Keller and

Schoene, 2012), coinciding with several major transition points in the Earth's geological evolution (e.g. the rise of atmospheric oxygen; Kump, 2008). While geochemical discrimination of (meta) basalts that formed in convergent versus divergent plate margin settings should ideally be applied in order to restrict these changes to MOR-derived units only (e.g. Furnes et al., 2014), there remains debate regarding how effectively trace element ratios used to discriminate modern-day mantle source regions can be applied in the geological past (Li et al., 2015). For example, assumptions that Archean basalts with high Nb/Th and Zr/Nb ratios may have had MOR-type affinity (e.g. Condie et al., 2016) stem from observations made from Phanerozoic data (Pearce, 2008), and a tectonically equivalent plate margin environment may not have been characterized by the same geochemical signatures on the early Earth.

Many geodynamic models suggest that subduction would have been relatively inefficient on a hotter Earth, if it operated at all, and that the early Archean may instead have been characterized by an active stagnant lid regime (O'Neill et al., 2007; Van Hunen and Moyen, 2012; Sizova et al., 2014). Recently, Condie (2015) showed that Archean basalts generally differ from Proterozoic and Phanerozoic basalts by having very few enriched- and depleted-mantle source signatures, as defined by Zr/Nb and Nb/Th ratios. Most data obtained from samples with MOR-type geological characteristics were tightly clustered around primitive mantle ratios, with some overlap into the hydrated mantle field. This pattern may indicate a relatively unfractionated stagnant-lid mantle source, which suggests that mantle convection accompanying this regime would have inefficiently recycled various enriched and depleted components in the deep mantle. If true, low-MgO basaltic rocks of Archean age may have initially formed by fractional crystallization processes rather than via closed-system partial melting (e.g., Foley et al., 2002, 2003). However, the petrological characteristics of basalts generated in a stagnant lid tectonic setting are very poorly understood, and applying arbitrary compositional ranges to geochemical databases as a predefined criterion to exclude particular lithologies (e.g. Gill, 1979; Condie, 1985; Condie et al., 2016) may inadvertently cause a false impression of compositional quiescence through time – at least in terms of overall maficity. New analytical data obtained from studies of other planetary bodies in our solar system may provide insight into this problem (Downs, 2015), as Mars, for example, may have been characterized by such a stagnant lid regime when most of its mafic crust formed. Recent investigation into the effects of metamorphism on such stagnant-lid versus plate-tectonic basalts (Wade et al., 2017) indicate notable petrological and petrophysical differences between the two for equivalent metamorphic conditions. This supports our interpretations that metamorphism of primitive crust in the Archean would have produced dramatically different rock types than observed today, even in intraplate tectonic settings.

What conclusions can thus be drawn about the petrological nature of Archean oceanic crust from studying basalts in the geological record? While some greenstone belts preserve rare examples of deformed and metamorphosed remnants of oceanic lithosphere (e.g. sheeted dykes: Helmstaedt et al., 1986; Kusky and Kidd, 1992; Furnes et al., 2007; Grosch and Slama, 2017), no complete sections of juvenile crust are known (Bickle et al., 1994). Evolved tholeiitic basalts with low MgO contents appear to represent major volumetric constituents of both Archean and Proterozoic greenstone belts (Condie, 1981), although these basalt compositions are not in equilibrium with mantle peridotite (O'Hara and Herzberg, 2002) and so do not themselves represent primary magmas. Primary mantle melts from a hotter mantle would have been more MgO-rich than today, and the lowermost portions of the Archean crust must have comprised large volumes of complementary cumulates significantly more mafic than those observed today (Fig. 1). While a defining characteristic of Archean greenstone belts is the presence of extremely MgO rich (>25 wt.%) komatiitic basalts (Condie, 1981), these lavas are typically interpreted to have formed via intraplate plume-related activity at elevated mantle T_P (Arndt et al., 1997), and thus cannot represent these hypothesized complementary residues to low-MgO tholeiites. Neither ultramafic cumulate-like lithologies nor their metamorphosed equivalents (garnet pyroxenite: Fig. 3) have been documented *en-mass* in Proterozoic or Archean terranes (Bickle et al., 1994; Bédard, 2017), and geophysical data do not suggest their presence at cratonic bases (Abbott et al., 2013). Petrophysical arguments have been made to suggest that such high-density residuum delaminated into relatively lower-density underlying aesthenospheric mantle (Vlaar et al., 1994; Van Thienen et al., 2004; Bédard, 2006; Fischer and Gerya, 2016), explaining their apparent absence from the geological record.

3.2. Implications for uniformitarianism and the onset of modern-day plate tectonics

The principle of uniformitarianism is succinctly summarized by the phrase "*the present is the key to the past*". Uniformitarianistic arguments have been used by many studies over the past few decades to investigate exactly when modern-day plate tectonics – defined by one-sided subduction – initiated on Earth, with propositions spanning almost the entirety of Earth history between the Hadean (>4.2 Ga; Hopkins et al., 2008) and the late Neo-Proterozoic (0.85 Ga; Hamilton, 2011). A wide variety of geological data and/or theoretical modelling results have been utilized to support the conclusions of each of these studies. Predicted and observed changes in oceanic crust/basalt compositions (Table 1) may be used to shed light on first-order petrological arguments based on lithologies in the geological record that form in or around subduction zone environments (Stern, 2005; Palin and White, 2016).

Rock types reported to be diagnostic of metamorphism along low-temperature/high-pressure geotherms that characterize subduction zones today include, but are not limited to, glaucophane- and lawsonite-bearing mafic rocks (blueschists), and ultrahigh-pressure (UHP) lithologies (coesite- and diamond-bearing eclogites). The rarity of these lithologies from the rock record prior to ca. 0.7 Ga was interpreted by Stern (2005) to argue for a Neo-Proterozoic onset of modern-day subduction-driven plate tectonics. However, the phase equilibria presented here for rocks of variable MgO and Al_2O_3 contents (Fig. 3) indicate that bulk composition exerts a strong control on the mineral assemblage that should form in mafic crust descending into either warm (Fig. 4a–d) or cool (Fig. 4e–h) Archean subduction zones.

Lawsonite-bearing assemblages are shown in our calculations to be stable in almost all mafic bulk compositions at low-grade conditions (Fig. 4e), but will only remain during continued metamorphism in relatively low-MgO lithologies (Fig. 4f), with more mafic protoliths forming garnet and clinopyroxene instead. The relative availability of H_2O and Na_2O will control whether glaucophane or talc additionally occurs. As such, lawsonite should not be expected to appear in subducted and metamorphosed MgO-rich basalt that likely typified the uppermost levels of Archean oceanic crust (Ganne and Feng, 2017), rendering its petrological significance for diagnosing the operation of subduction throughout Earth history dubious. Furthermore, even lawsonite that is known to have formed at blueschist- and eclogite-facies conditions in Phanerozoic subduction zones commonly does not survive the exhumation process (Clarke et al., 2006; Whitney and Davis, 2006), and thus its absence in Precambrian orogens may be strongly influenced by preservation bias. We further note that lawsonite's breakdown products typically include chlorite, zoisite, paragonite, and/or quartz (Ballevre et al., 2003; Groppo and Castelli, 2010; St-Onge et al., 2013), and so its retrograded products may superficially resemble assemblages observed in greenstones that dominate Archean terranes (cf. Palin and White, 2016).

Similarly, the absence of coesite- or diamond-bearing UHP mafic rocks cannot necessarily be used as an *a-priori* indicator of the non-operation of subduction, as many eclogite-facies rocks subducted during Proterozoic and Phanerozoic orogenesis record maximum pressures below the quartz–coesite transition (~26–28 kbar; Palin et al., 2014; Möller et al., 2015; Weller et al., 2015, 2016; Maldonado et al., 2016). However, the lower silica content of highly magnesian basalts (Table 1; Arndt et al., 1997) may prohibit the formation of quartz/coesite within the metamorphic assemblage, even if UHP conditions are reached (cf. Klein et al., 2017), highlighting a further limitation of applying the modern-day facies definitions to Archean rocks. Given that both lawsonite-bearing blueschists and UHP eclogites are absent from many convergent margins today, they should be viewed as sufficient—but not necessary—indicators of subduction-related processes on the early Earth, and their absence may instead be accounted for by secular change in MOR-derived basalt composition, or else a dramatic change in exhumation processes at the Proterozoic–Phanerozoic boundary (Korenaga, 2016). Therefore, modern-day facies classification systems that are fundamentally built around mineral assemblages that stabilize in low-MgO metabasalt compositions cannot be confidently applied to ancient metamorphic terranes without considering this bulk-rock composition effect.

4. Conclusions

Thermal models of the Earth's internal heat budget predict that the Archean mantle was notably hotter than the present day (Herzberg et al., 2010), which would have resulted in higher degrees of partial melting during mantle decompression, and the formation of a thicker and more magnesian oceanic crust (Van Thienen et al., 2004). The absence of pristine examples of obducted ophiolites in the geological record precludes direct examination of the geochemistry of primary magmas or their MOR-derived basalts, which would validate such theoretical predictions; however, numerous recent studies have attempted to use statistical methods to examine the degree of secular compositional change from global datasets of metamafic rocks. These works corroborate the theoretical predictions, and suggest that early Archean basalts may have contained nearly ~15 wt.% MgO, compared to just over ~7 wt.% for modern-day examples (White and Klein, 2014). However, wide compositional variation occurs in modern-day basalts according to variation in tectonic setting and mantle potential temperature of formation (e.g. Iceland: Winchester and Floyd, 1977). If such variation also existed in the geologic past, mean compositions of basalts of various ages would reflect biases in sampling from various tectonic settings. Overall compositional changes in basalts with time would thus be obscured. Classifying the geodynamic environments of formation of Archaean and Proterozoic mafic igneous rocks is therefore critical, although it is uncertain how well trace element signatures diagnostic of particular geodynamic environments of formation on the modern-day Earth may translate into the geological past.

What are the implications of a secular change in oceanic crust/MORB composition? A longstanding discussion exists in the geoscience community as to why modern-day petrological indicators of subduction, such as lawsonite-blueschists and UHP metamorphic rocks, suddenly appear in the geological record at ca. 0.7 Ga (e.g. Stern, 2005; Brown, 2007) if subduction on a global scale has operated since at least the Meso-Archean (~3.0 Ga). We have analyzed phase assemblages that would stabilize during metamorphism of variably mafic basalts, and show these diagnostic rock types may not form in high-MgO protoliths potentially representative of the upper levels of Archean oceanic crust (Fig. 4). This suggests that the modern-day metamorphic facies system, which is predicated on the mineral assemblage changes that occur in low-MgO mafic protoliths, must be applied to rocks in Archean terranes with great care. It should therefore be remembered that '*the absence of evidence is not evidence of absence*'. Multi-disciplinary studies that are able to link multiple independent lines of evidence into holistic tectonic models, such as recently demonstrated by Wade et al. (2017), are required to make significant progress in this field in the future.

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